RADIOLOGICAL SURVEY OF AN ESTATE CONTAMINATED BY THORIUM FOR REUTILIZATION PURPOSES

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Summary

A measuring method for the radiological survey of an estate contaminated by natural thorium has been developed and applied by the combination of the following three nuclear measuring procedures of different capacity, accuracy and sensitivity: surface contamination measurement on the spot; integral γ and β intensity measurement in a mobile measuring station using sampling technique; instrumental γ-spectrometry in the basic laboratory.

Introduction

Owing to the widespread use of radioactive isotopes environmental pollution hazards have been increased significantly. The major tasks of health and environmental protection involve the prevention of contamination and the decontamination of sites which were contaminated as a consequence of an accident or of human negligence. Decontamination procedures can be efficient and economic only if the spatial distribution and the degree of contamination were determined previously. Different measuring methods must be combined with one an other, to meet the partially contradictory demands of high sensitivity, accuracy and capacity on the contamination mapping.

The radiological survey of an estate where chemical activities involving the application of thorium salts used to be run was carried out to measure the contamination levels of natural thorium compared to the maximum permissible level thus determining the conditions of a possible reutilization.

Different types of surfaces of a total area of about 1500 m^2 were to be monitored including soil and lawn in the garden, wall, ceiling and flooring surfaces in the house. Plant samples from the garden were investigated, too. Dividing all the surfaces into squares (rasters) of 1 m × 1 m — hence assuring the reliability of the survey — the contamination level of about 1500 spots had to be measured. The actual methods selected for the measurements were primarily determined by the great number of samples. Regarding the aspects of

sensitivity and accuracy as well, the combination of the following measuring methods and devices was chosen:

— high resolution γ-spectrometry using semiconductor detector and MCA (conducted in our nuclear laboratory);
— integral γ and integral β intensity measurement using a combined γ, β scintillation device;
— in situ surface contamination measurement using a portable γ scintillation equipment.

Selection of the measuring methods (Fig. 2)

Natural thorium and its daughter isotopes emit α, β and γ rays (Fig. 1). Because of the great number of samples the only reasonable way to measure the activity of ^{232}Th was the methodologically simple γ detection (self absorption of the samples can be neglected) with some additional γ measurements.

1. The most exact (but not the most effective) method of measuring ^{232}Th activity — based on the detection of γ-radiation — is γ-spectrometry. Knowing the detection efficiences, the isotopic abudances and the decay scheme of ^{232}Th the activity and the amount of the parent nuclide can be determined by means of the γ-spectra. γ-spectrometric measurements give the opportunity to control the radionuclide content sample by sample in order to avoid the error caused by the possible segregation of the elements or by the transient state of the secular equilibrium. The capacity of γ-spectrometry due to the time-consuming measurement and data evaluation is very low; 2 samples/day, making the

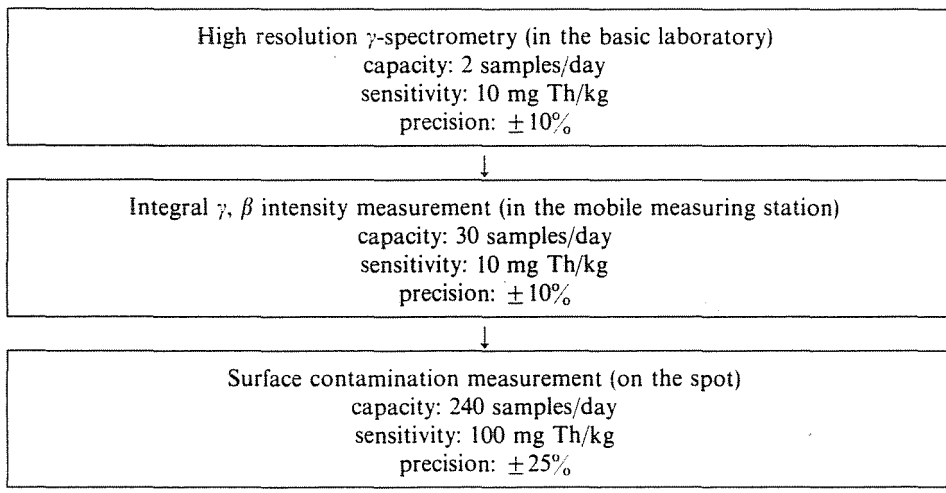

Fig. 1. Block scheme for the selection of the measuring methods

stand-alone γ-spectrometric contamination survey of the whole estate impossible. The application of this technique was restricted to the determination of thorium concentration in some control samples, but it helped increase the reliability of the results obtained by the integral γ intensity measurements.

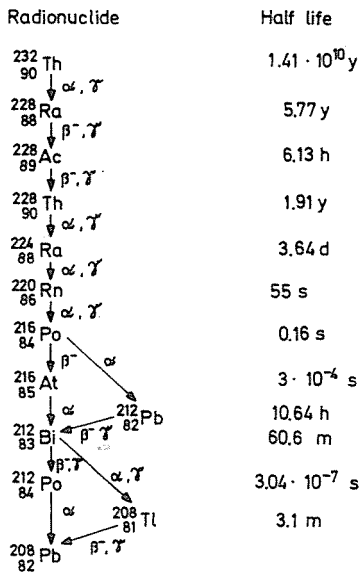

Fig. 2. Decay chain of natural thorium

2. ^{232}Th concentrations can be calculated from the measurement of integral γ-intensity data of collected samples by means of a proper calibration standard of the same age as that of thorium in the sample. (The isotope ratios of thorium descendants i.e. the age of the samples should be controlled occasionally by γ-spectrometry.) The capacity of the method using this sampling technique (30 samples/day) is still too low to measure 1500 samples during an acceptable time interval.

Integral β intensities can be measured in the same samples and thorium concentrations calculated by a similar calibration method making use of β, γ combined scintillators.

3. The radiological survey of the whole area can be carried out by a portable surface contamination meter. Counting rates must be determined on the spots. The capacity of this method is high enough to carry out 240 measurements daily but its accuracy is low due to the γ-radiation of the neighbouring rasters. To assess the concentration of ^{232}Th the counting rates of surface contamination measurements were calibrated with the calculated

concentrations of the same raster position determined by the integral γ measuring method.

Combining the measuring methods described above the radiological survey of the whole estate could be carried out within some weeks with the acceptable precision of 25% on average.

Measuring devices

γ-spectrometer:

γ-spectra were taken with a Ge(Li) detector and a 4k MCA "ICA–70" type. Data were evaluated by an on-line HP 9825/A desk top calculator (1).

energy resolution: 3.2 keV/1332 keV
energy range: 100–1600 keV with a 0.8 keV/channel gain
photopeak efficiency: 1–0.08%.

Mobile measuring station (2):

The integral γ and β intensities of the samples were determined by a γ, β sandwich scintillator (plastic — NaI(Tl)). The measuring vessel of a total volume of 0.9 dm^3 was designed according to "Marinelli" geometry. Pulse signals were evaluated by a two-channel energy selective analyzer (NP–357).

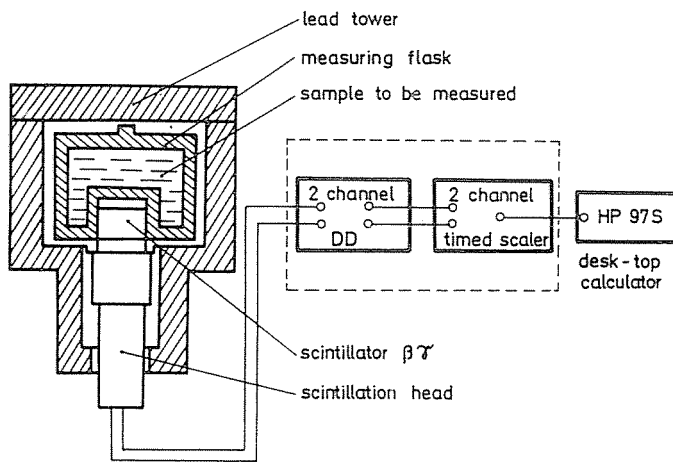

Fig. 3. The block scheme of the mobile measuring station

The measuring station is operated in a microbus. The block scheme of the measuring system is shown in Fig. 3.
energy range: 100–2000 keV
counting efficiency: $>4\%$
detection limit: 10–100 Bq/dm^3

Portable γ-counter:

The signals of the NaI(Tl) scintillator were acquired by a portable scaler "NC–484". The detector was placed on the surfaces of interest.
energy range: >20 keV
sensitivity: the counting rate of a ^{226}Ra source of a dose rate of 0.01 μGy/h was 12 cps

Results and discussion

Gamma-spectrometry

The γ-spectra of some samples contaminated to a greater extent were taken to determine the isotope composition of the samples. According to the measurements the following conclusions could be drawn:

— The samples did not contain γ-emitting radioactive isotopes other than the members of the natural thorium chain.
— Analysing the intensity ratios of the full energy peaks, it could be established that the activity ratios of the radionuclides of the thorium chain are in good agreement with the values calculated from the decay scheme, i.e. isotope enrichment of segregation was negligible.
— Because of the lack of isotope enrichment the theoretical radioactive concentration of thorium could be calculated to be 4031 ± 205 Bq/g ^{232}Th. (For the calculations the existence of the decay equilibrium was supposed.)
— The actual radioactive concentration of thorium was determined by taking the γ-spectra of known amounts of thorium salts. It was found to be 3681 ± 413 Bq/g ^{232}Th, so comparing the measured and the calculated values a good agreement of 10% accuracy (inside the one standard deviation range of the measurement) was observed, proving that the quasiequilibrium of the secular decay chain had been attained.

Because of its low capacity γ-spectrometry was applied only to control the isotope composition and the existence of secular equilibrium in a limited number of samples.

In order to confirm the assumed insolubility of thorium compounds solvent extraction measurements were carried out with some soil samples using either distilled water or distilled water saturated with carbon dioxide. No

significant radioactivity could be detected in the solutions hence supporting our concept on the immobility of thorium compounds in soil. (The same conclusions can be drawn from the fact that no radioactivity could be detected in the collected plant samples either.)

Integral γ, β intensity measurements

The thorium concentration of more than one hundred of samples were determined based on integral γ and β intensity measurements in the mobile measuring station. The counting efficiency of this method is the highest due to the application of scintillation detector and "Marinelli" geometry. Thorium concentrations were calculated by a standard calibration method using thorium-nitrate of about the same age as that of the thorium compounds contaminating the estate. The results of concentration determinations based on γ and β intensity measurements, respectively, showed good agreement. The sensitivity of the method was about 10 mg Th/kg soil, the estimated standard deviation — including the inhomogeneity of the samples — was $\pm 15\%$.

A simple gamma-ray logging measurement was carried out by the same method using the sampling technique to determine the distribution of thorium in the function of depth from ground level (an important question regarding the possible decontamination). The maximum of thorium concentration was found to be at about 30 cm below the ground level.

Surface contamination measurement

Contamination mapping of the whole estate could be carried out by the fast surface contamination measurements only. The surface contamination at more than thousand spots was determined using the portable γ counter. To calculate the approximate thorium concentration at the given rasters a calibration curve was taken plotting the thorium concentrations calculated from the integral γ intensity measurement as the function of the γ counting rate measured for the same raster of the estate (Fig. 4). A straight line was fitted to 105 measured points, the goodness-of-fit was found to be sufficient ($=0.90$). Naturally this method has the lowest sensitivity (100 mg of Th/kg of soil) and precision (the standard deviation was $\pm 25\%$) due to the disturbing radiation of the neighbouring rasters.

To illustrate the results of the radiological survey a part of the plan of the estate is given in Fig. 5 showing the thorium concentration distribution determined by this method.

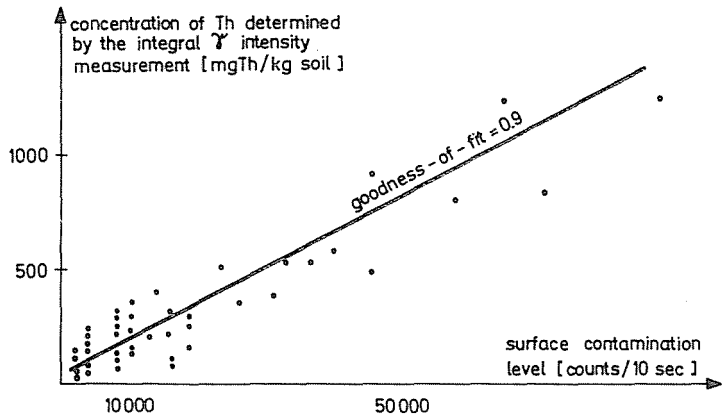

Fig. 4. Calibration line to determine Th concentration by means of surface contamination measurements

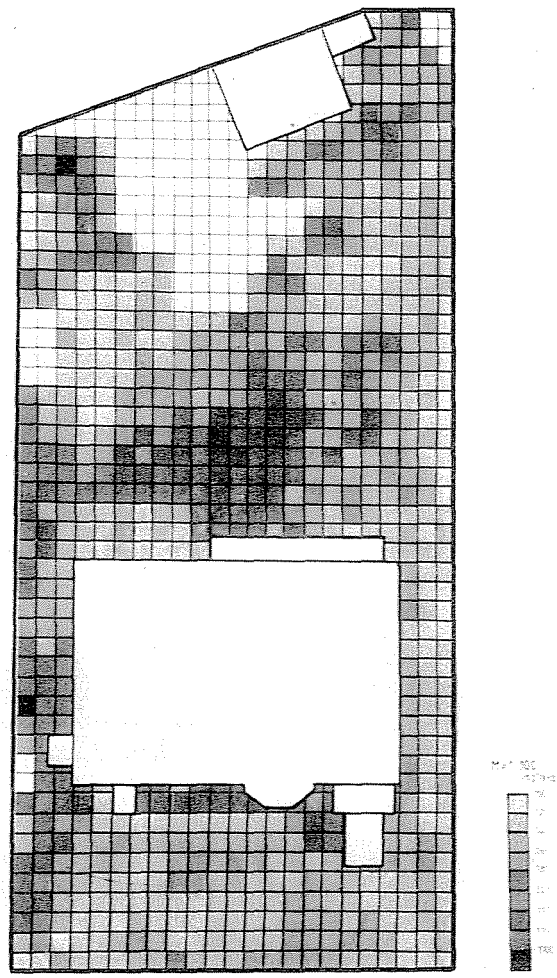

Fig. 5. Radiological survey of the estate

References

1. ZAGYVAI, P.: Computerized evaluation of γ-Spectrometric measurements Dissertation, 1982
2. SZABÓ, K.–NAGY, L. GY.–TÖRÖK, G.–HEGEDÜS, D.–FÓTI, G.: Method and instrumentation to monitor radioactive concentrations in waste waters J. Radioanal. Chem., *58*, 393 (1980)
3. Hungarian standard: MSZ 19384–81 Transportable measuring station for rapid in situ determination of radioactivity in liquid samples (1981)

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